^{224}Ra – Comments on evaluation of decay data
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Evaluation Procedures

Limitation of Relative Statistical Weight Method (LWM) was applied to average numbers throughout the evaluation. The uncertainty assigned to the average value was always greater than or equal to the smallest uncertainty of the values used to calculate the average.

Decay Scheme

A relatively simple decay scheme was constructed from the alpha-particle studies of 1962Wa28, 1969Pe17, 1971So15 and 1984Bo15, and the gamma-ray measurements of 1969Pe17, 1972DaZA, 1977Ku15, 1982Sa36, 1983Sc13, 1983Va22, 1984Bo15, 1984Ge07 and 1992Li05. Only the gamma-ray studies of 1977Ku15 provide any detail beyond the 240.986 keV gamma ray; all other measurements are dedicated to the determination of the absolute emission probability of the 240.986 keV gamma ray. A weighted mean emission probability was determined for this transition, and the other emission probabilities as measured by 1977Ku15 were subsequently adjusted.

Cluster decay has been observed by 1985Pr01 and 1991Ho15, and reviewed by 1995Ar33 and 1997Tr17. ^{14}C emissions were detected with a branching fraction of 5 (1) 10^{-11}. However, this decay mode has not been included in the decay-data summary section.

Nuclear Data

^{228}Th decay chain is important in quantifying the environmental impact of the decay of naturally-occurring ^{232}Th. Specific radionuclides in this decay chain are noteworthy because of their decay characteristics (^{224}Ra alpha decay to ^{220}Rn; ^{212}Bi and ^{208}Tl gamma-ray emissions).

Half-life

The recommended half-life represents the least-squares weighted mean of two somewhat elderly studies (1962Ll02 and 1971Jo14) and a much more recent measurement (2004Sc04). Further measurements are required to determine this half-life with greater confidence.

Reference	Half-life (d)
1962Ll02	3.62 (1)
1971Jo14	3.665 (38)
2004Sc04	3.6319 (23)
Recommended value	3.631 (2)

There is no evidence of any change in the half-life of ^{224}Ra on extreme cooling of ^{224}Ra samples within a metallic environment (2007St23). Sources were held at temperatures at and below 1 kelvin for periods of several days, and exhibited an upper limit of change in the alpha-decay half-life of the order of 1 %.

Alpha Particles

Energies

All alpha-particle energies were derived from the structural details of the proposed decay scheme. While the energies of the two main alpha-particle emissions have been directly measured by 1962Ba19, 1962Wa28, 1971Gr07 and 1991Ry01, the nuclear level energies of 1997Ar04 and evaluated Q-value of 5788.85 (15) keV (2003Au03) were used to determine the recommended energies and uncertainties of the alpha-particle emissions, and by allowing for the significant recoil components.

Adopted nuclear levels of ^{220}Rn: J^{π} and origins (1997Ar04).

Nuclear level	Nuclear level energy (keV)	J^{π}	Origins
0	0.0	0 +	^{220}At β^- decay, **^{224}Ra α decay**
1	240.986 ± 0.006	2 +	^{220}At β^- decay, **^{224}Ra α decay**
2	533.69 ± 0.10	4 +	^{220}At β^- decay, **^{224}Ra α decay**
3	645.44 ± 0.09	1 –	^{220}At β^- decay, **^{224}Ra α decay**
4	663.03 ± 0.10	(3 –)	^{220}At β^- decay, **^{224}Ra α decay**

Measured and recommended energies of the main alpha-particle emissions of ^{224}Ra.

	E_α (keV)				
	1962Ba19	**1962Wa28**	**1971Gr07**	**1991Ry01**	**Recommended value***
$\alpha_{0,1}$	5447.2 (12)	5447	–	5448.6 (9)	5448.80 (15)
$\alpha_{0,0}$	5684.0 (12)	5684	5685.56 (20)	5685.37 (15)	5685.48 (15)

* Determined from the nuclear level energies of 1997Ar04 and evaluated Q-value of 5788.85 (15) keV (2003Au03).

Emission Probabilities

The alpha-particle emission probability to the first excited state of ^{220}Rn has been directly measured by 1969Pe17, 1971So15, 1984Bo15 and 1993Ba72. These data were also used to calculate the alpha-particle emission probability directly to the ground state of ^{220}Rn:

The emission probability data of 1969Pe17 for the 5685.48- and 5448.80-keV alpha particles have been effectively normalized to 94.95 (5) % and 5.05 (5) %, similarly for the equivalent data of 1971So15 with normalized values of 95.1 (4) % and 4.9 (4) %, and 1984Bo15 with normalized values of 94.94 (4) % and 5.06 (4) %. Equivalent relative emission probabilities measured by 1993Ba72 also require normalization to give P_α(5448.80 keV) of 4.93 (4) % and P_α(5685.48 keV) of 95.10 (4) %. These alpha spectrometry measurements by 1969Pe17, 1971So15, 1984Bo15 and 1993Ba72 can be used to determine a weighted mean P_α(5685.48 keV) of 95.00 (4) % (0.9500 (4)) that can be matched with a value of 5.01 (4) % (0.0501 (4)) for P_α(5448.80 keV). However, these data were not adopted – both the measurement and spectral analysis techniques used to determine the gamma-ray emission probabilities were judged to be more reliable, and therefore preference was given to alpha-particle emission probabilities derived by calculation from the recommended gamma-ray emission probabilities and their theoretical internal conversion coefficients.

Alpha-particle emission probabilities per 100 disintegrations of ^{224}Ra.

E_α(keV)	P_α						
	1953As31	**1962Wa28**	**1969Pe17**	**1971So15**	**1977Ku15#**	**1984Bo15**	**1993Ba72**
5034.29 (18)	-	0.0031	-	-	0.0029 (5)	-	-
5051.56 (17)	-	0.0072	-	-	0.0073 (10)	-	-
5161.32 (18)	-	0.0073	-	-	0.0069 (8)	-	-
5448.80 (15)	4.9	5.5	5.05 (5)	4.9 (4)	[5.00 (16)]	5.06 (4)	[4.93 (3)]¶
5685.48 (15)	95.1	94	94.95 (5)	95.1 (4)	[94.98 (16)]	94.94 (4)	[95.1 (6)]¶

\# Data were deduced from gamma-ray studies.

¶ Data are relative, and were adjusted to P_α^{abs} (5685.50 keV) of 95.10 (4) and P_α^{abs} (5448.80 keV) of 4.93 (4).

Alpha-particle emission probabilities per 100 disintegrations of ^{224}Ra, and hindrance factors.

E_α(keV)	P_α						HF
	1969Pe17	1971So15	1984Bo15	1993Ba72	LWM value#	Recommended value*	
5034.29 (18)	-	-	-	-	-	0.0030 (5)	6.4
5051.56 (17)	-	-	-	-	-	0.0076 (10)	3.7
5161.32 (18)	-	-	-	-	-	0.0072 (8)	17.9
5448.80 (15)	5.05 (5)	4.9 (4)	5.06 (4)	4.93 (4)	5.01 (4)	5.25 (5)	1.04
5685.48 (15)	94.95 (5)	95.1 (4)	94.94 (4)	95.10 (4)	95.00 (4)	94.73 (5)	1.00

Limitation of relative statistical weight method applied to the measured alpha-particle emission probabilities, with the uncertainty adjusted from ± 0.03 to ± 0.04 so as not to fall below the lowest measured uncertainty.

* Recommended alpha-particle emission probabilities derived from evaluated gamma-ray emission probabilities and theoretical internal conversion coefficients.

There is an unsatisfactory lack of agreement between derivations of the decay scheme by means of the measured gamma-ray emission probabilities, compared with an equivalent procedure involving the measured alpha-particle emission probabilities:

(i) assuming that the measured gamma-ray emission probabilities are absolute (as quoted in the various references), the LWM of P_γ(240.986 keV) is 0.0412 (4), NF = 1.000, and balanced population-depopulation of the 240.986-keV nuclear level of ^{220}Rn provides a means of determining P_α(5685.48 keV) of 0.9473 (5):

P_α(5448.80 keV)

$= P_\gamma(240.986\ \text{keV})[1 + \alpha_{tot}(240.986\ \text{keV})] - [\Sigma\ P_{\gamma i}(1 + \alpha_i)\ \text{populating nuclear level}]$

$= [0.0412\ (4) \times 1.276\ (4)] - 0.000\ 124\ (11) = 0.0524\ (5)$,

and P_α(5685.48 keV) = 0.9473 (5).

(ii) if an absolute least-squares weighted mean value for P_α(5685.48 keV) of 0.9500 (4) is adopted from the alpha-particle measurements of 1969Pe17, 1971So15, 1984Bo15 and 1993Ba72, NF = 0.95 (5) and P_γ(240.986 keV) is 0.0392 (4) (also defined as 3.92 (4) %).

The measured gamma-ray emission probability data were judged to be more reliable – this important decision is based on the assumed superiority of the gamma-ray spectroscopic procedures and spectral analysis techniques in the 1970s/1980s. Thus, the recommended alpha-particle emission probabilities were determined from the gamma-ray data and theoretical internal conversion coefficients, rather than the available alpha-particle measurements. These gamma-ray calculations resulted in an absolute emission probability of 0.0525 (5) for the 5448.80-keV alpha particle (compared with a least-squares weighted mean value of 0.0501 (4) from the alpha-particle measurements that represents a difference of 4.6 %), and 0.9473 (5) for the 5685.48-keV alpha particle (compared with a least-squares weighted mean value of 0.9500 (4) from the alpha-particle measurements that represents a difference of only 0.3 %). Although not recommended, note is also made that combining the two different sets of data results in weighted means of 0.0513 (12) for P_α(5448.80 keV), and 0.9487 (13) for P_α(5685.48 keV).

A hindrance factor (HF) of 1.00 for the 5685.48-keV alpha-particle emission yields r_0(^{220}Rn) of 1.5421 (1), which was adopted in the equivalent HF calculations for the other alpha emissions.

Gamma Rays

<u>Energies</u>

All gamma-ray transition energies were calculated from the structural details of the proposed decay scheme. The nuclear level energies of 1997Ar04 were adopted, and used to determine the energies and associated uncertainties of the gamma-ray transitions between the various populated-depopulated levels.

Emission Probabilities

Absolute emission probabilities were determined from measurements of the 240.986-keV gamma ray by 1969Pe17, 1972DaZA, 1982Sa36, 1983Sc13, 1983Va22, 1984Bo15, 1984Ge07 and 1992Li05. A weighted mean value of 4.12 (3) % was derived through LWEIGHT, and the uncertainty was increased slightly to the lowest measured value of ± 0.04 to give 4.12 (4) % (0.0412 (4)).

Only 1977Ku15 has measured the emission probabilities of other low-intensity gamma transitions identified with ^{224}Ra alpha decay; these data are reported relative to a value of 39 500 (1 300) for the 240.986-keV gamma emission, as taken from 1969Pe17. Hence, the low-intensity emission probabilities have been subsequently adjusted on the basis of P_γ(240.986 keV) of 4.12 (4) % (0.0412 (4)).

Absolute gamma-ray emission probabilities per 100 disintegrations of ^{224}Ra.

E_γ (keV)	P_γ^{abs}				
	1969Pe17	1972DaZA‡	1977Ku15†	1982Sa36	1983Sc13
240.986 (6)	3.95 (13)	3.9 (7)	[3.95 (13) → 4.12 (4)]	3.9 (2)	4.04 (17)
292.70 (10)	-	-	0.0063 (7)	-	-
404.45 (9)	-	-	0.0022 (5)	-	-
422.04 (10)	-	-	0.0030 (5)	-	-
645.44 (9)	-	~ 0.007	0.0054 (9)	-	-

E_γ (keV)	P_γ^{abs} (cont.)				
	1983Va22	1984Bo15	1984Ge07	1992Li05	Recommended value*
240.986 (6)	4.05 (9)	4.05 (9)	4.17 (4)	4.11 (12)	4.12 (4)
292.70 (10)	-	-	-	-	0.0063 (7)
404.45 (9)	-	-	-	-	0.0022 (5)
422.04 (10)	-	-	-	-	0.0030 (5)
645.44 (9)	-	-	-	-	0.0054 (9)

‡ Data expressed relative to P_γ(2614.511 keV) of ^{208}Tl have been adjusted.
† Data adjusted on the basis of P_γ(240.986 keV) of 4.12 (4) %.
* Recommended gamma-ray emission probabilities above 241 keV taken from adjusted data of 1977Ku15.

Multipolarities and Internal Conversion Coefficients

The nuclear level scheme specified by 1997Ar04 has been used to define the multipolarities of the gamma transitions on the basis of known spins and parities. All of the recommended internal conversion coefficients have been determined from the frozen orbital approximation of Kibédi *et al*. (2008Ki07), based on the theoretical model of Band *et al*. (2002Ba85, 2002Ra45). Uncertainties of ± 1.5 % were adopted for all of the E1 and E2 gamma transitions.

Gamma-ray emissions: multipolarities and theoretical internal conversion coefficients (frozen orbital approximation).

E_γ (keV)	Multipolarity	α_K	α_L	α_{M+}	α_{total}
240.986 (6)	E2	0.110 9 (16)	0.122 0 (17)	0.043 1	0.276 (4)
292.70 (10)	E2	0.072 7 (11)	0.056 4 (8)	0.019 6	0.148 7 (21)
404.45 (9)	E1	0.014 01 (20)	0.002 41 (4)	0.000 75	0.017 17 (24)
422.04 (10)	(E1)	0.012 80 (18)	0.002 19 (3)	0.000 68	0.015 67 (22)
645.44 (9)	E1	0.005 46 (8)	0.000 894 (13)	0.000 276	0.006 63 (10)

Atomic Data

The x-ray and Auger-electron data have been calculated using the evaluated gamma-ray data, and atomic data from 1996Sc06, 1998ScZM and 1999ScZX. Both the x-ray and Auger-electron emission probabilities were determined by means of the EMISSION computer program (version 4.01, 28 January 2003). This program incorporates atomic data from 1996Sc06 and the evaluated gamma-ray data.

K and L X-ray emission probabilities per 100 disintegrations of ^{224}Ra.

			Energy (keV)	Photons per 100 disint.
XL		(Rn)	10.137 – 17.280	0.373 (16)
	XL_l	(Rn)	10.137	0.007 74 (20)
	XL_α	(Rn)	11.598 – 11.726	0.138 (4)
	XL_η	(Rn)	12.855	0.004 13 (11)
	XL_β	(Rn)	13.520 – 14.565	0.191 (5)
	XL_γ	(Rn)	16.770 – 17.280	0.042 4 (9)
XK_α	$XK_{\alpha2}$	(Rn)	81.07	0.130 (3)
	$XK_{\alpha1}$	(Rn)	83.78	0.214 (4)
$XK'_{\beta1}$	$XK_{\beta3}$	(Rn)	94.247	)
	$XK_{\beta1}$	(Rn)	94.868	) 0.074 3 (18)
	$XK''_{\beta5}$	(Rn)	95.449	)
$XK'_{\beta2}$	$XK_{\beta2}$	(Rn)	97.48	)
	$XK_{\beta4}$	(Rn)	97.853	) 0.023 8 (7)
	$XKO_{2,3}$	(Rn)	98.357	)

Electron energies were determined from electron binding energies tabulated by Larkins (1977La19) and the evaluated gamma-ray energies. Absolute electron emission probabilities were calculated from the evaluated absolute gamma-ray emission probabilities and associated internal conversion coefficients.

Data Consistency

A Q_α-value of 5788.85 (15) keV has been adopted from the atomic mass evaluation of Audi *et al.* (2003Au03) while in the course of formulating the decay scheme of ^{224}Ra. This value has subsequently been compared with the Q-value calculated by summing the contributions of the individual emissions to the ^{224}Ra alpha-decay process (i.e. α, electron, γ, etc.):

$$\text{calculated Q-value} = \sum (E_i \times P_i) = 5788.7\ (40)\ \text{keV}$$

Percentage deviation from the Q-value of Audi *et al.* is (0.00 ± 0.07) %, which supports the derivation of a highly consistent decay scheme.

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